RECOMBINATION RATES AND NON-EQUILIBRIUM ELECTRICAL CONDUCTIVITY IN A SEEDED PLASMA

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Abstract

Experimentally determined values of electrical conductivity and electron temperature have been measured in a non-equilibrium seeded plasma. These results are in good agreement over a wide range of parameters with values calculated from a two-temperature model of the plasma. There is no doubt that the two-temperature model is valid over a wide range of gas temperatures, seed concentrations, and current densities for the argon-potassium and helium-potassium plasmas. However, the model does not give an accurate description of the plasma when the current density is below about 0.4 amp/cm^2; in this range the omission of the influence of atom-atom excitation and the influence of non-equilibrium excited state populations may explain the discrepancy between experiment and theory. In addition, the electron-electron-ion collisional recombination rate for potassium has been measured in the argon-potassium system. The range of electron temperatures investigated was between 1900° K to 3000° K with electron densities between 3×10^{13} and 4×10^{14}/cm^3. The measured values show a scatter of 60 per cent about theoretical values calculated from present recombination-rate theory employing the Gryzinski classical collision cross sections.

Introduction

The work reported in this paper is the result of an investigation of certain properties of plasmas made up of a noble gas seeded with potassium vapor. The primary purpose of this work has been the experimental determination of the electric conductivity of the gas and the verification of the two-temperature model for the calculation of the conductivity as a function of electric field or current density. The key assumption of the two-temperature model is that in the presence of an electric field, electrons and excited states of ~~neutral particles~~ the heavy species are in thermal equilibrium at an elevated temperature and that the ~~neutral particles~~ heavy species are in translational equilibrium at a lower temperature. As a result of the elevated electron temperature, the electron density and hence the conductivity will be much greater than that existing in the equilibrium state at the ~~neutral gas~~ heavy species temperature. In the model, the temperature difference between the two systems composing the plasma is fixed by the balance between the energy input to the electrons and the excited states from the electric field, and the energy loss from this system due to elastic energy transfer to the ~~neutral particles~~ heavy species and radiant energy transfer from the excited states.

In a previous paper[1], the authors have presented a theoretical treatment of this problem which was a development of the original work of Kerrebrock[2] and others[3,4]. This treatment allows the determination of electrical conductivity, the electron temperature, and electron density of the plasma as a function of the current density, the plasma composition and temperature, and the elastic electron momentum transfer cross sections for the species present in the plasma.

An experimental program has been carried out to obtain values of the conductivity and to determine the validity of the proposed model and the calculations. The most easily measured characteristic of the plasma is the electrical conductivity, and this is the quantity which has been studied most thoroughly. However, under the conditions studied here, the electrical conductivity is very roughly proportional to the current density, and hence the agreement of calculated and measured values does not give a precise check on the model. For example, if one calculates conductivity as a function of current density and assumes that the electron density is only half that obtained in the two-temperature model, no measurable difference in the curve of conductivity versus current density is obtained. The only measurable difference is that the calculation would predict a 10 per cent higher electron temperature for a given current density. Hence to check the validity of the proposed model, direct measurements of electron temperature and number density are necessary.

In the present paper, the results of the previously reported conductivity measurements are briefly reviewed, and new data extending the measurements to the very low current-density range are presented. A discussion is then given of electron temperature measurements, and these results are compared with the calculated values.

In the final section of the paper, experimental measurements and calculations of the electron - electron-ion collisional recombination rates for potassium are presented and are compared with each other.

Experimental Apparatus

The basic apparatus used in the experiments has been described in detail elsewhere[1] and only a brief description is given here. An arc-jet heater, shown schematically in Figure 1, is used to heat the larger portion of a noble gas flow. This main flow is then combined with a smaller, secondary flow of the noble gas which has been saturated with potassium vapor by passing it through a potassium boiler with a minimum residence time of 2 sec. The combined flow passes through a mixing chamber with a length-to-diameter ratio of 16 before entering the test section. Measurements of recombination rates in the plasma indicate that this length is more than enough to allow complete relaxation of the plasma to steady conditions. The concentration of potassium in the flow, which results from the combination of these two streams, is fixed by controlling the boiler temperature and secondary gas rate. Measurements indicate that the potassium concentration in the resulting stream is within

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10 per cent of the calculated flow rates.

A typical test section is shown in Figure 2. Electrons are emitted thermionically from a spiral tungsten electrode immersed in the flow at the downstream end of the cylindrical test chamber and flow axially to the anode that is a stainless steel cylinder imbedded in the insulating wall. The test section inside diameter was 0.460 in.; typical dimensions of the other elements are shown in Figure 2 in units of inches. Tungsten-wire voltage probes of 0.040-in. diameter were inserted as shown. It was established that the voltage gradient along the duct was constant by placing as many as 6 probes in the flow.

The insulating cylinder used as the test-section wall was made of boron nitride, and it was thermally shielded so that the inner wall temperature was above 1250° K. The two sets of ports shown in Figures 1 and 2 were used in making spectral intensity measurements. The upstream set was equipped with a cell which could be purged with an auxiliary flow of argon gas. This prevented the condensation of potassium on the quartz windows as well as preventing the flow of the test gas through the port. Without this port, a cloud of cool potassium vapor was formed in the optical path which gave absorption effects. Previous difficulties in temperature measurements[1] were traced to the presence of this cloud which lead to measured temperatures below the true value. The auxiliary argon flow rate used to purge the ports was kept below $1\frac{1}{2}$ per cent of the flow rate of the test gas and had no observable effect on measured values of the conductivity. In the recombination experiments, the port through which the plasma was viewed was shielded in a similar manner by the light pipe which connected the port and the photomultiplier tubes.

The physical arrangement of the photomultiplier tubes and the spectroscope used in the temperature determination experiments is shown in Figure 1. This spectroscopic equipment and associated electric circuits are conventional. The rise time for pulses observed with the photomultiplier tubes was less than a microsecond. The tubes were cooled to dry ice temperatures to decrease the noise level, and condensation in the optical system was prevented by purging with dry, high-purity nitrogen.

Photomultiplier tubes and optical filters were selected to isolate various regions of the spectrum containing three spectral transitions of potassium and one in sodium. The tubes and filters used to monitor each transition are shown in the following table:

Table I.

Transition	Bausch and Lomb 2nd Order Interference Filter Number	Kodak Wratten Filter Number	RCA Phototube Number
K_I 4P→4S	33-79-76	89B	7102
K_I 5P→4S	33-79-40	0	7102
K_I 6S→4P	33-78-69	29(F)	6655-A
Na_I 3P→3S	33-79-58	22	7102

In order to make as precise light intensity measurements as possible for the steady state measurements of temperature, it was found necessary to use electronic filters to eliminate high frequency noise from the photomultiplier tube output. The use of these filters enabled relative intensity comparisons to be made with an estimated error of less than 2 per cent. Conventional shielding arrangements were used throughout the experimental work to eliminate possible spurious pick-up of electric signals.

The optical equipment was arranged so that it was possible to make as many as three simultaneous spectral observations. By rotation of a mirror, it was also possible to use the same port to make steady state measurements of the SLR temperature. The SLR apparatus was calibrated with an optical pyrometer which in turn was calibrated against a NBS standard lamp. Calibration of the apparatus was carried out with great care and followed the standard procedures described in Ref. 5.

The distribution of neutral gas temperature in the test gas was determined by observing the temperature of a graded series of fine tungsten wires inserted at various radii in the test duct. The diameters of these wires ranged in size from 0.012 to 0.003 inches. The wire temperatures were determined by use of an optical pyrometer and were corrected to the true wire temperature through use of the standard emissivities for tungsten. The gas temperature was deduced from the wire temperatures through the standard techniques discussed in Ref. 6. Typical data and a calculated gas temperature profile are given in Figure 4. The dashed line gives the bulk or mean temperature of the gas, averaged over the cross section of the duct. This is the temperature referred to as the gas temperature, T_a.

Electrical Conductivity

Measurements of conductivity of an argon-potassium seeded plasma have been made over a wide range of parameters[1,7]. The measurements were made at a pressure of one atmosphere and at neutral gas temperatures between 1250° K and 2000° K, current densities between 0.4 and 90 amp/cm^2, and at potassium concentrations between 0.1 and 1.0 mole per cent. In addition, measurements were made at a single test condition with a helium-potassium seeded plasma.

In addition to this experimental work, values of the conductivity for these systems were calculated from a two-temperature model[1,7]. Included in this model were the energy dependence of the electron-atom momentum transfer cross sections, the effects of electron-ion encounters on the electrical conductivity and on the energy transfer from the electron gas, and also the effects of radiant energy loss from the whole system. Although the model is based on the assumption that the electron distribution function is Maxwellian to the first approximation, no undetermined constants appear in the model, and once the cross sections of the plasma species are given, the conductivity and the electron density and temperature can be directly calculated as a function of current density.

The agreement between the calculated and measured values is illustrated by the data pre-

sented in Figures 5 and 6. For both figures, the data are in excellent agreement with the calculated values when the effects of radiant energy loss are properly included in the calculations and when current densities greater than $\frac{1}{2}$ amp/cm^2 are considered. This agreement is typical of that obtained over the whole range of parameters for which data were previously reported[1, 7]. The fact that good agreement is obtained for gases with electron - atom momentum transfer cross sections as different as argon and helium indicates that the general computational scheme is correct. In addition, note that a wide range of current densities is covered in these experiments. At current densities near 1 amp/cm^2 in the argon-potassium system, electron-ion interactions are negligible compared to electron-atom encounters; in contrast, at current densities near 80 amp/cm^2, the Coulomb encounters are predominant. Thus, the good agreement between experiments and theory indicates that the use of the Spitzer conductivity expression and the inclusion of electron-ion energy exchange in the theoretical calculations of Ref. 1 appears to be substantially correct.

In previous papers[2, 7], Kerrebrock and the present authors have reported that at very low current densities the conductivity values reached a plateau and remained constant with decreasing current. In most cases, the constant value was considerably above the equilibrium value of the conductivity. This anomalous behavior has been investigated by the authors, who have found that in their apparatus, at least, it is a result of a thin, poorly-conducting film which builds up on the walls of the test section and which apparently shunts the voltage probes. This film appears to the eye to be a shiny surface which covers the entire test-section wall. If tests are carried out with new boron nitride test sections before this film appears, low current-density measurements can be made which do not show the plateau. Values obtained with a "dry" wall are shown in Figure 5; the new data correspond to the range $.025 \leq J \leq 0.40$ amp/cm^2. In earlier work[7], during which the wall was apparently slightly conducting, the plateau conductivity was about 0.2 mho/cm, and it began at about 0.4 amp/cm^2. In contrast, the conductivities shown in Figure 5 decrease in a smooth way toward the equilibrium value. The authors feel that the data presented here are correct and that the previous results were the result of the shunting effect.

These data are of interest from another point of view. Note that in the neighborhood of 0.2 amp/cm^2 the experimental values break away rather sharply from the theoretical curve which includes the effect of radiant energy loss from the whole system. There are a number of possible reasons for this deviation. The most plausible explanation, to the authors, is connected with the energy balance for the first few excited states which account for the majority of the radiant energy loss. For high electron density, the excitation of these states by electron-atom interactions and the self-absorption of the radiation is sufficient to keep the population density of these states in thermal equilibrium with the free electrons. However, as the electron density falls, the rate of excitation by electron-atom interactions will fall, and at low enough electron density the population levels will no longer be kept in thermal equilibrium with the free electrons. Under these conditions, calculated values of radiant energy loss will be too large.

A second and perhaps more important effect is that there is an essentially constant rate of excitation of these low-lying states by atom-atom interactions. Hence, at low enough electron density, the atom-atom interaction will become the dominant excitation mechanism and the population levels of the excited states will be more nearly in thermal equilibrium with the neutral particles than with the electrons.

Both of these processes lead to a situation in which too large a radiant energy loss is calculated; for this situation, the calculated conductivities should be less than the measured values. This simple picture of the energy balance also indicates that, in the limit of zero current density, the excited states may have population levels corresponding to temperatures somewhat lower than the neutral gas temperature due to radiant loss effects.

The low current results also throw some light on a practical problem. Examination of the data of Figure 1 indicates that the field strength increases monotonically with current density but that between 0.4 and 1.5 amp/cm^2 the field strength is almost constant at about 2 volts. In this range, the current density and conductivity are very sensitive to small changes in field strength. Similar unstable regions were observed at other potassium concentrations in argon- and helium-potassium experiments. The unstable region is produced by the large influence of the radiation correction in this low current range.

In the unstable range, $d\ln\sigma/d\ln J$ is approximately unity, and consequently the field strength is almost constant. However, if the radiation correction had been larger, as it would be in a smaller apparatus, the value of $d(\ln\sigma)/d(\ln J)$ would increase above unity and consequently the field strength would have a local maximum in this region. Such a maximum in the local field strength is indeed observed by Kerrebrock and Hoffman[2], who worked in an apparatus with a diameter about 60 per cent of that used in the present experiments. If the foregoing explanation is correct, this maximum is a result of the more important radiation loss in their smaller system and will not exist in a larger device in which radiation effects will be much less important.

An interesting phenomenon has been reported by other authors[8, 9] in connection with similar experiments both in argon-potassium and helium-cesium plasmas. Two modes of current conduction have been observed. The first of these is encountered at low current densities, and under this condition the plasma emits a uniform diffuse glow from the entire volume. However, at a higher current density, a transition is observed to a second mode of conduction in which the discharge constricts to a relatively narrow, arc-like discharge. The critical value of the current apparently depends on the temperature of the plasma and the seed concentration. The authors have never observed the second type of behavior in their experiments. Visual observation indicates that the discharge always gives rise to a diffuse uniform glow which fills the entire cross section of the duct and which increases in intensity with current density.

In summary, the agreement between the calculated and measured values of electrical conductivity appears to be excellent over a wide range of parameters for argon-potassium and helium-potassium plasmas as long as the current density is above a value of about $\frac{1}{2}$ amp/cm^2. Below this current density, the calculated radiation loss is too large and the calculated conductivity is too low. The principal error in the calculations appears to be the neglect of the atom-atom mode of excitation of the low-lying electronic states.

Electron Temperature Measurements

The good agreement between calculated and measured values of the electrical conductivity shows that the two-temperature model is useful for making conductivity estimates but does not provide a very precise check of the key assumptions made in the model. It was felt that a good check could only be obtained by making a more direct measure of electron temperature or density. In this section, a description is given of the experiments used to obtain a measure of the electron temperature. The recombination experiments described in the following section were used to obtain estimates of electron density.

Two spectroscopic techniques were used to determine population "temperatures" of electronic states of neutral sodium and potassium atoms. In order to obtain the electron temperature from these data, it is necessary to show that these states are in thermal equilibrium with the electron gas. Calculations for the argon-potassium system of the total probability of electronic collisional de-excitation and also the total probability of radiative decay, including the effects of absorption, were made for the low-lying levels of potassium up to and including the 4F level at an electron temperature of 2600° K and an electron density of 10^{14}/cm^3. The results showed that the probability of radiative decay is always less than one hundredth of that for collisional de-excitation by electrons. Hence it seems reasonable to expect that for this electron density the excited state and the electron gas are in equilibrium. For higher excited states, equilibrium is even more likely. Note that for a current density of 2 amp/cm^2 the calculated value of electron density in the plasma under study is greater than 10^{14}. For current densities below 2 amp/cm^2 the electron density drops off, and calculations indicate that the equilibrium assumption becomes questionable for current densities around $\frac{1}{2}$ amp/cm^2. In summary, it appears to be reasonable to assume that thermal equilibrium does exist between the electronic excited states and the electron gas when current densities above 2 amp/cm^2 or electron densities above 10^{14} are used. Hence, in this range it is also reasonable to equate population temperatures and electron temperature.

The first spectroscopic technique used to determine a population temperature was the sodium line reversal or SLR method which utilizes the 3P→3S transition of neutral sodium. This technique is well known, e.g., Ref. 5, and provides an absolute measure of the population of the 3P level and hence an absolute measure of the population temperature. However, since the light source used in the experiments was a tungsten strip lamp, the measured values were limited to temperatures below 2950° K.

The second technique was a relative method based on the use of the change in light intensity resulting from a given transition in response to a change in the applied electric field. The intensities of the 3P→3S transitions of neutral sodium, and the 4P→4S and 5P→4S transitions of neutral potassium were measured before and after an electric field was applied to the plasma.

Since the intensity of a transition is proportional to the population density of the upper state, one can determine the population temperature of the upper state by measuring this intensity. The absolute intensity observed depends in detail on absorption of the plasma and the apparatus used to measure the intensity. However, relative changes in intensity can be used to infer relative changes in population temperature, provided the absorption of the line does not change.

For the conditions of the present test, the line spreading is due to Stark and collision broadening as well as Doppler broadening. However, calculated values of the Stark broadening are negligible compared to the other two effects, and thus changes in electron density should have no effect on absorption. In addition, resonance lines were employed, and thus slight changes in number density of atoms in the ground state have a negligible effect for the conditions discussed here. Consequently, the absorption does not change as a function of current or electron density. In the absence of changes in absorption, the intensity ratio for a given line can be written as

$$\frac{I\{T_2\}}{I\{T_1\}} = \exp\left[\frac{\Delta E_i}{kT_1} - \frac{\Delta E_i}{kT_2}\right] . \qquad (1)$$

Here, I is the intensity of the ith transition, T is the temperature, k is Boltzmann's constant, and ΔE_i is the energy difference of the ith transition. The subscript 1 refers to conditions when the electric field is zero, and 2 to conditions when a field is applied. This equation can be solved for the temperature ratio to give:

$$\frac{T_1}{T_2} = 1 - \frac{(kT_1)}{\Delta E_i} \ln\left(\frac{I_2}{I_1}\right) . \qquad (2)$$

Equation (2) was used to obtain temperatures from intensity data for the 3P→3S transition in neutral sodium. The measurements were made up to temperatures of about 3000° K, and for these calculations the value used for T_1 was obtained from the SLR measurements. These data and that obtained from the SLR technique are shown as the solid points in Figure 7. The two sets of data agree reasonably well in the region between 6 and 8 amp/cm^2 where they overlap. This indicates that the relative method and the use of equation (2) is consistent with the SLR method and gives confidence in the relative method as it is used here.

In addition to the sodium measurements, data were obtained with two transitions in potassium. However, in order to use equation (2) with this data, it is necessary to determine the values of the temperatures in the zero field case. This is a non-trivial problem, because radiation losses keep the various excited states out of thermal equilibrium and because no absolute technique such as the SLR method was readily available. The method used to obtain the initial temperatures was as follows. Relative intensity data were obtained simultaneously for the 3P→3S

sodium transition and the 4P→4S potassium transition. The value of T_1 for the potassium transition was picked to minimize the squared deviations between T_2 values calculated from equation (2) for the two transitions. (This calculation is based on the assumption that the various lines will be in thermal equilibrium when the electron density is high.) In another set of experiments, simultaneous measurements of the 4P→4S and 5P→4S potassium transition intensities were made. The value of T_1 for the 5P→4S was picked to minimize the squared deviations between the two sets of data.

The zero field temperatures calculated by this technique for an 1800° K temperature of the 3P→3S sodium transition were 1790° K for the 4P→4S potassium transition and 1865° K for the 5P→4S potassium transition.

Use of these values of zero field population temperatures for the two potassium lines gives the values of population temperature shown in Figure 7 as the open points. The data agree very well with each other and with the values obtained from the sodium data for the whole range of current densities investigated. This good agreement indicates that the equilibrium assumption made in interpreting the intensity data is justified.

In Figure 7, brackets have been placed on some of the SLR data and on some of the potassium data to indicate the maximum errors. The maximum uncertainty in the SLR data is about $\pm$ 40° K, and this uncertainty is due to errors in technique and drift in operating conditions for the arc-jet heater. The larger uncertainty for relative measurements reflects this $\pm$ 40° K maximum uncertainty in the zero-field SLR temperature for these data.

The 10° K difference between the population temperatures of the sodium and 4P→4S potassium transition is well within the accuracy of the techniques, and the close agreement in these temperatures is to be expected from the similarity of the transitions in the two atoms. The higher temperature obtained for the 5P→4S line is significant, since detection of a 65° difference is within the capabilities of the technique. This difference indicates that the system is not in thermal equilibrium in the zero-field case and suggests that the electron temperature is somewhat higher than the temperature of the lowest excited state. Note that the mean temperature for the neutral gas, for the conditions under which these data were obtained, was about 2000° K. (Gas temperature measurements are discussed in a previous section.) If these measurements are correct, the zero-field temperatures of the low-lying states of potassium are about 200° K (4P→4S) and 135° K (5P→4S) below the mean temperature of the neutral gas.

The solid curve shown on Figure 7 represents the calculated value of electron temperature for the experimental conditions used in these experiments. Although the data are somewhat scattered, they clearly fall along the curve and hence are in excellent agreement with the calculated values. As was pointed out earlier, there is good reason to expect that for currents of 2 amps/cm^2 or higher, all of the excited states studied were in equilibrium with the electron gas. Hence, the comparison of measured and calculated temperatures, shown in Figure 7, does constitute a check on the electron temperature calculation. It is evident that the calculation gives a satisfactory estimate of electron temperature except at very low current densities.

Recombination Rates

The remainder of this paper deals with measurements which were made of electron-electron-ion recombination rates and with their comparison with values calculated from a model utilizing the classical inelastic collision cross section expressions of Gryzinski. Following a discussion of the measurements, the energy balance and recombination rate calculations are presented. After establishing this theoretical background, the data reduction technique is given and the results are discussed.

Recombination Measurements

The apparatus and electric circuit used to measure recombination rates are shown in Figures 1 through 3. The measurement technique consisted of first establishing a steady initial discharge in the argon-potassium plasma. The field strength was then abruptly reduced to a very low value, and during the relaxation period immediately following the reduction of the electric field, measurements were made of the electrical conductivity and the relative populations of several of the excited states of potassium. In a later section it is shown that it is possible to deduce the electron recombination rates from these measurements.

The range of current densities used in the initial discharge was between 2 and 17 amp/cm^2, which corresponds to field strengths of about 2.5 and 5 volts/cm and to initial electron densities of about 1.5 and 8×10^{14}/cm^3. The electric field strength was kept above zero during the relaxation period so that the conductivity of the gas could be monitored. The final field strengths used were between 0.3 to 0.8 volt/cm.

A typical set of experimental data is reproduced in Figure 8. Here, light intensity data from two consecutive tests are presented as a function of time. Also shown on the figure are the probe voltages used in obtaining the conductivity values and the values of the electron density calculated from the conductivity values. The transient in the applied field dies out within two microseconds and the probe voltage difference remains essentially constant thereafter.

Light intensity data are shown for the 5P→4S and 4P→4S transitions in K_I. The 5P→4S light intensity initially decays slightly more rapidly than the electron density and the 4P→4S light intensity, reaching a relatively low value by about 20 μsecs, and changes more slowly thereafter. A similar behavior was noted for the non-resonance 6S→4P transition of K_I (not shown). Interpretation of data for the latter transition must include consideration of the effect on absorption due to changes in the populations of the 4P level with time.

Note in Figure 8 that the measurements of the decay of the light intensities have been made for nearly identical initial conditions, but for final conditions both with and without a small applied electric field. The values for the 4P→4S transition appear slightly displaced in the vertical direction. This reflects the small random fluctuation in initial

light intensity commonly observed and is of no importance in interpreting the results. The important thing to note is that the relative changes in population for the 4P and 5P levels appear to be nearly identical with or without an applied field. The effect on the decay rates due to the presence of the small field (0.39 volt/cm for this case) is negligible and shows that the relatively small energy input to the free electrons by this field during the relaxation process probably has no important effect on the free-electron recombination rate.

Also note the initial lag in the decay of the first excited state population (4P→4S light intensity) which is probably due to the initial downward cascading from higher states to the 4P level, as will be discussed at the end of the following sections.

Energy Balance

In order to analyze the recombination data, one must have some means of determining the electron temperature during the relaxation process. The electron temperature and electron density are not independent quantities since an energy balance on the free electrons defines their instantaneous temperature for a given instantaneous electron density. During the initial steady state before the drop in electric field has been initiated, the electron temperature is defined by a balance between energy input from the electric field and energy loss due to elastic collisions as well as inelastic collisions. As has been discussed in Ref. 1, inelastic energy losses are accounted for in the steady case by calculating the radiant energy loss from the excited states. This calculation gives the amount by which losses due to electronic collisional excitations and ionizations are not completely balanced by the energy input due to electronic de-excitations and recombinations, provided that radiative recombination is negligible.

After the abrupt reduction of the electric field, the electron energy loss terms temporarily exceed inputs to the free electrons and cause the electron temperature to decrease. As the electron temperature falls, the rates of recombination and de-excitation exceed those of ionization and excitation. For the electron densities of interest here, the electrons can exchange energy with the heavy species by elastic and inelastic collisions in times much shorter than those associated with changes in the population of the free electrons. This results in the rapid establishment of a quasi-steady condition in which the instantaneous electron temperature is given by an energy balance. The important energy inputs are a result of electronic collisional recombinations and de-excitations and the acceleration of electrons by the small remaining applied field. The important energy losses are due to ~~the acceleration of electrons by~~ electronic excitations and ionizations, and to elastic collisions with the neutral and ionized heavy species.

During this quasi-steady relaxation period it is meaningful to ascribe a temperature to the free electrons, provided that the density of electrons is high enough to provide mutual electronic interactions sufficient to keep the electron energy distribution Maxwellian. The general criteria for the existence of a Maxwellian distribution of free electron energies are probably satisfied for the conditions of these experiments[9, 10]; however, its existence will be postulated rather than completely justified.

The quasi-steady energy balance may be written for the conditions of the relaxation experiments described here as

$$\sigma E^2 - (V_o+\tfrac{3}{2}kT_e)\frac{dN_e}{dt} - \left(\sum_i V_i \frac{dN_i}{dt} + \dot{R}\right) = \dot{\Omega}. \quad (3)$$

In equation (3), the net rates of energy input per unit volume to the free electrons have been written on the left hand side of the equation; these balance the elastic energy losses on the right hand side. σE^2 is the rate of energy gain by the free electrons from the small remaining applied field. The remaining terms on the left hand side represent the net energy gain by the free electrons as the result of collisional recombination and de-excitation, and the collisional decay of the populations of the various excited states (all d/dt terms are negative during decay). The ionization potential of potassium is given by V_o and is 4.34 ev. The quantity $-\sum_i V_i(dN_i)/dt$ is the net rate at which energy is transferred to free electrons due to changes in the populations, N_i, of the excited states, i. Here, V_i is the energy of the ith level as measured relative to the ground state. If the populations of the excited states are also quasi-steady in addition to the electron energy, then the $(dN_i)/dt \approx 0$. $\dot{R}$ represents the total rate per unit volume at which radiation from bound-bound transitions is lost from the system. The $\dot{\Omega}$ term represents the rate at which free electrons lose energy due to elastic collisions with both neutral and ionized species, and has been presented in Ref. 1. Note that the formulas in Ref. 1 can be easily modified to include other systems with atomic species of widely differing masses.

Equation (3) neglects the possibility of direct radiative recombination as will be justified in the following sections.

Calculation of Recombination Rates

With the recent development of the classical collision cross-section expressions of Gryzinski[11], it has been possible to formulate expressions for the rate constants for the electronic collisional processes occurring in high-density plasmas similar to the ones studied here. These rate expressions have led to theoretical electron-ion recombination formulations utilizing a model which includes recombination to excited levels by electron-electron-ion collisions, electronic collisionally-induced transitions between excited levels, as well as the radiative transitions between bound states. Careful experimental work has established a remarkably good agreement between experimental results and theory for such differing atomic species as hydrogen, helium, and cesium.[12-14] In addition, detailed theoretical calculations have been performed giving recombination rates for argon and potassium.[15]

There is no need to discuss here the details of the theoretical calculations performed by these several authors and others, but the physical model for the recombination process deserves brief comment with regard to the principal assumptions and approximations used in order to relate experiments and theory.

As mentioned in the previous section, the electron temperature in high-density plasmas quickly reaches a quasi-steady value given by an instantaneous energy balance on the free electrons. A similar, and in fact directly coupled, phenomenon exists with regard to the population densities of highly excited states in the recombining plasma. The detailed classical formulation of electronic inelastic collisions indicates that the probability of a collisionally-induced transition between two neighboring atomic levels is inversely proportional to the square of the energy gap separating them. Thus, the number densities of those states most closely spaced adjust rapidly with the changes in electron temperature. In the recombining plasma, this means that the populations of those upper states nearest the continuum tend to come into a mutual quasi-steady collisional equilibrium with the free electrons; and in addition, closely spaced lower levels reach a mutual quasi-steady equilibrium in times that are small compared to the time scale for changes in the populations of the free electrons and the ground state. This fact has resulted in an enormous simplification in the solution of the coupled rate equations describing the rate of change of the number density of the free electrons as well as the population of the infinite number of excited levels. The result has been the reduction of an infinite set of coupled differential equations to a finite set of algebraic equations describing the rate of change of the population of the free electrons and the ground state. Further requirements for this simplification, including that the population of a given level be much less than the free electron density in order for that level to be regarded as having a quasi-steady population, are discussed in detail by Bates, et al.[14]

The important physical result is that the upper levels and the free electrons tend to be in a quasi-steady mutual collisional (Saha) equilibrium, while the quasi-steady populations of the lower levels must be described by including the effects of radiative transitions between levels. Bates, et al. have performed detailed calculations for hydrogen and helium using this method, and the resulting rates and population densities have been confirmed experimentally be several authors.

Byron, et al.[15] have shown that good results can be obtained with fairly simple calculations by treating the collisional recombination process as a chain of reactions in which the net rate of recombination can be described in terms of the slowest step in the chain. The physical picture consists of a reservoir of upper excited states in quasi-steady collisional equilibrium with the free electrons above a critical energy gap across which recombining atoms must pass to reach the ground state. The critical energy gap will be the one of all possible energy gaps across which the total de-excitation rate is a minimum. A minimum occurs since the probability of de-excitation of a given level will increase with increasing principal quantum number of the level due to more closely spaced gaps, while the populations of the various levels decrease with increasing principal quantum number. In addition, the total probability of radiative decay of a given level decreases with increasing quantum number.

The Byron formulation has the powerful advantage of being easily applied to atoms with arbitrary energy level structures which are not necessarily hydrogenic. The authors have here used this calculation technique to obtain recombination rates for potassium in the range of interest of these experiments which occur at slightly higher electron temperatures than the maximum temperatures in the published results of Bryon, et al.[15] As justified in Ref. 15, the rate of crossings in the upward direction across an energy gap U located above a level of binding energy E_1 and coming from all initial energy levels with binding energies E_i, $(E_i \geq E_1)$, to all possible final bound energy states as well as the continuum can be written as

$$\left.\frac{dN}{dt}\right|_u = \sum_i \left.\frac{dN_i}{dt}\right|_u = \sum_i N_i N_e \bar{c} \frac{\sigma_o B_i}{(U+E_i-E_1)^2}\left(1+\frac{2kT_e}{U+E_i-E_1}\right) e^{-\frac{U+E_i-E_1}{kT_e}}$$

and writing

$$N_i = \frac{N_1 g_i}{g_1} e^{\frac{E_i-E_1}{kT_e}},$$

$$\left.\frac{dN}{dt}\right|_u = N_e\left(\frac{N_1}{g_1} e^{-U/kT_e}\right)\bar{c}\left[\sigma_o \sum_i \frac{B_i g_i}{(U+E_i-E_1)^2}\left(1+\frac{2kT_e}{U+E_i-E_1}\right)\right].$$

where $B_i = B_i\{E_i/(E_i-E_1+U)\}$ is the slope of the Gryzinski cross section g_i as given in Figure 2, Ref. 15; $\sigma_o = 6.56\times10^{-14}$ cm^2, and $\bar{c}$ is the mean thermal speed of electrons, $\sqrt{(8kT_e)/\pi m_e}$. N_1 and g_1 are the number density and degeneracy, respectively, of the energy level with binding energy E_1. N_e and T_e are the free electron density and temperature, respectively.

The summation is to be taken over all energy levels below the gap $(E_i \geq E_1)$ and avoids the necessity of assuming the levels below the gap to be continuous, as is done in equation (2), Ref. 15.

At equilibrium, the total rate of crossing the gap in the upward direction must balance the total rate of crossings in the downward direction. Also, at equilibrium, $N_2/N_1 = (g_2 \exp(-U/kT_e)/g_1)$ where N_2 and g_2 are the number density and degeneracy, respectively, of the energy level with binding energy E_1-U located immediately above the gap. Thus, at equilibrium,

$$\left.\frac{dN}{dt}\right|_d = \frac{(N_2 \text{ equil})}{g_2} N_e \bar{c} \sigma_o \left[\sum_i \frac{B_i g_i}{(U+E_i-E_1)^2}\left(1+\frac{2kT_e}{U+E_i-E_1}\right)\right] . \quad (4)$$

The minimum value of the total rate of downward crossings, $dN/dt|_d$, found by calculation from equation (4) for each energy gap, can be related to the recombination rate. At this point, some difficulties are encountered. Equation (4) gives the total rate of crossings of the gap in the downward direction at collisional (Saha) equilibrium. However, to find the recombination rate to the ground state, one must subtract from

this rate such downward crossings between bound states above and below which are the result of atoms that are de-excited to a lower level, but instead of being eventually further de-excited to the ground state, are again re-excited to a level above the gap. An additional complication is the fact that the actual recombination rate may be somewhat less than the total de-excitation rate calculated at equilibrium, since N_2 may not in general be equal to its equilibrium value, $N_{2\ equil}$.

Byron, et al.[15] have investigated these effects and have found that $dN_e/dt = \gamma(dN/dt)|_d$, where γ lies between 1/4 and 1. The latter value applies when the minimum is very pronounced, as at very high temperatures, and decreases to 1/4 if the minimum is not well defined, as at lower temperatures. For the temperature range of these experiments, $\gamma \cong 1/3$.

Calculations of $dN/dt|_d$ were performed for the low-lying levels of potassium to establish that for electron temperatures between 2000° K and 3000° K, the minimum de-excitation rate occurs for the gap lying below the lumped 6S and 4D levels and above the 5P level. Resulting values of the recombination rate coefficient, $1/N_e^3 (dN_e/dt)$, calculated with $\gamma = 1/3$ for $2000^\circ K \leq T_e \leq 3000^\circ K$ are shown in Figure 10. The recombination rate coefficient calculated at 2000° K agrees with the value at 2000° K in Ref. 15. Also shown in Figure 10 are the values for cesium as calculated in a more exact fashion with an extensive program by Dugan[16]. Note that the calculated rates for cesium and potassium are nearly equal, which would be expected, judging from the similarity of their energy level structures.

At no point in the foregoing calculations have the effects of radiative transitions upon the recombination rate appeared explicitly. Such radiative transitions can cause non-equilibrium populations in the levels immediately above the gap. This effect should be considered in addition to the non-equilibrium effects resulting from the fact that quasi-equilibrium may not have been established because of the relatively long times required for this condition to be established in low-lying states. The factor γ could be interpreted to include all non-equilibrium effects, including radiative de-population; however, one must still include the possibility of direct radiative recombination when calculating the total rate of recombination. The radiative recombination rate for hydrogenic atoms can be written[17] as: $\alpha_r \cong 2.7 \times 10^{-13}/(kT_e)^{3/4}$ cm^3/sec, and the radiative recombination rate for potassium should not be substantially different. Thus, at $T_e = 2500^\circ$ K, $kT_e \approx 0.215$ ev and we find $\alpha_r \cong 9 \times 10^{-13}$ cm^3/sec. Measured values of $\alpha = -1/N_e^2 (dN_e/dt)$ from these experiments are of the order of 10^{-10} cm^3/sec, and hence direct radiative recombination appears to be negligible.

The effects of diffusion of ions to the walls as well as recombination by the dissociation reaction $K + K \rightleftharpoons (K_2^+) + e^-$ are assumed negligible for the densities and temperatures of these experiments. Estimates of the mobility of K^+ in K and K^+ in A, the former based on that for Cs^+ in Cs,[18] indicate diffusion losses to be small compared to measured recombination rates. The very small concentrations of the molecular ions at these temperatures and seed concentrations make dissociative recombination improbable. Harris[19] has reached similar conclusions based on his calculations and measurements in cesium for conditions of temperatures and densities approximating those encountered here. Finally, it should be mentioned that the inert gas plays no appreciable role in the ionization and recombination processes studied here, due to its relatively high ionization potential and the relative inefficiency of atom-atom excitations and ionizations.

Data Reduction

The directly measured quantities during the relaxation period immediately following the abrupt reduction in field strength are the electric field and current density as well as the relative populations of the various transitions in potassium which were observed spectroscopically. From these quantities, we compute the electrical conductivity as a function of time and the number densities in the upper states of the observed transitions. The latter are obtained from the measured relative changes in population and from the measured initial conditions.

In order to determine the electron density and electron temperature, one must solve the equation for the quasi-steady energy balance on the free electrons given in equation (3) simultaneously with the expression for the non-equilibrium electrical conductivity. The conductivity expressions have been presented in detail in Ref. 1 and it is not necessary to repeat them here.

The calculation was carried out numerically using an iterative scheme. The first step was to obtain an initial estimate of the value for the electron density from the measured instantaneous value of the conductivity. In making this calculation, it was found that a good first approximation could be obtained by using the steady state relationship between the non-equilibrium conductivity and the electron density. Given this estimate for the electron density and the measured values of the relative populations of the excited states, it is possible to solve the quasi-steady electron energy balance equation for the electron temperature. This value for the electron temperature is then used to obtain a more accurate value for the electron density from the conductivity expression, and the process is continued until suitable convergence is achieved. Usually, one or two iterations are sufficient for the accuracy desired in these calculations. The various terms in quasi-steady energy balance equation must be evaluated from the experimental data. The evaluation of these terms is discussed in the following paragraphs.

Detailed calculations of the radiant energy losses from bound states of potassium at steady state for the argon-potassium system have been performed by the authors and are discussed in Ref. 1, and they are presented in more detail in Ref. 20 . These calculations show that about 70 per cent of the total radiation loss is accounted for by the $4P \rightarrow 4S$ transition in K_I for temperatures of 2000 to 3000° K. Thus, in the quasi-steady relaxation case, the measurement of the relative population of the 4P state in time allows a calculation of the radiation loss from the 4P state, since the initial steady-state intensity and radiation loss are known. The remaining, less than 30 per cent , of the radiation loss can be determined

if the general behavior of the relaxation of the various higher contributing transitions is known. This is afforded by the observation of the 5P→4S transition.

Calculation of the contributions of the various terms to the sum, $\Sigma V_i (dN_i/dt)$, show that the only term of appreciable magnitude is that of the 4P level, whose variation in population has been determined as stated above. This fact is due to the relatively large populations of the 4P level. Finally, calculation of σE^2 is done directly from the instantaneous experimental data. In this manner, instantaneous values of all the terms in equation (3), except that for $\dot{\Omega}$, are calculated directly from the experimental data, and thus equation (3) can be used to determine the variation of $\dot{\Omega}$ with time during the relaxation period. The first approximation to the instantaneous electron temperature can be obtained from values of $\dot{\Omega}$ by use of the equation:

$$\dot{\Omega} = (8/3)(m_e/m_a)(\epsilon_e - \epsilon_a)N_e \Sigma \bar{\nu}_m .$$

Here the $\bar{\nu}_m$ are the energy-averaged collision frequencies between the electrons and the neutral or ionized atoms in the plasma. Calculation of these terms is described in full detail in Ref. 1. The terms ϵ_e and ϵ_a represent $\frac{3}{2} kT_e$ and $\frac{3}{2} kT_a$, respectively.

The results of calculations for the intermediate set of data given in Figure 9 are shown below in Table II. Here, the various terms in equation (3) are evaluated for a number of times during the relaxation period. The last three rows of the table give the final values for the electron density and temperature, and the recombination coefficient, $\alpha \equiv (-1/N_e^2)dN_e/dt$. Values of the collisional recombination rate coefficient, $(-1/N_e^3)dN_e/dt$, are shown in Figure 10 as a function of the electron temperature. The data taken from Table II appear as the solid circles, and other data obtained from a number of different tests are also included in this figure.

Discussion

The values of the recombination coefficient calculated for times greater than 20 microseconds after the beginning of the relaxation are shown as the dark data points in Figure 10. These data are in fair agreement with the calculated points (open circles), although they are scattered by about 60 per cent for the range investigated.

The agreement is at least as good as one would expect when considering the possible uncertainty in the estimated values of the electron temperatures and those uncertainties connected with the theory. In particular, note that the theoretical rates are directly proportional to the constant γ, which could easily be 50 per cent higher or lower than the value used here. The theory is probably good to within a factor of 2 judging by the agreement of experimental measurements in cesium with calculated values[16], and the classical cross sections used are thought to be valid to within about a factor of 2. The principal uncertainties in the estimated electron temperatures are the result of possible errors in the gas temperatures as well as errors in the electron energy balance. These possible errors give a probable uncertainty in the data themselves of about a factor of two.

The half-filled data points of Figure 10 are those calculated for times as close to the time origin as possible, and in reducing this data it has been assumed that the electron temperature is at its initial value. Though these data would not be expected to show good agreement with the calculated values, since the quasi-steady populations did not have time to develop, it can be seen that fair agreement is obtained. At the other end of the temperature scale lie data obtained some 100 microseconds after the initiation of the relaxation process. Here, the electron temperatures are close enough to the gas temperatures to reduce substantially the uncertainties in electron temperature. Thus, the data for temperatures in the 1900 to 2300° K range are probably relatively more accurate.

Note that the 60 per cent scatter in the collisional recombination rate coefficients about the theoretical values would only correspond to a 30 per cent variation in electron density, even if all the scatter were due to errors in calculated electron density. Given this good agreement between experiment and theory, one can conclude that the electron densities as given by the two-temperature model are reasonable estimates of the actual plasma electron densities. Further, the recombination rates measured here for potassium appear to be in good agreement with present electron-electron-ion recombination rate theory.

The results given in Table II illustrate a difficulty encountered in determining the electron temperature with the quasi-steady energy balance as written in equation (3). The calculations have been performed starting backward from 140 microseconds after the voltage decrease was initiated and seem to give reasonable electron temperatures until about 20 microseconds. From this point back to the time origin, the calculated electron temperatures appear to be unreasonably high, and as the origin is approached closely, the calculated values exceed the measured initial temperature of 3000° K for this example. Thus, it appears that a true quasi-steady condition does not exist until after times of about 20 microseconds and that the population levels take at least this much time to distribute themselves in a true quasi-steady fashion.

Thus the term $-(V_o + \frac{3}{2} kT_e)dN_e/dt$ is probably too high to be a valid estimate of the rate of energy input to the free electrons during this early phase, since most of the electrons will recombine first into the uppermost levels and then these levels must be further collisionally de-excited to the ground state. Hence, the average rate of energy transfer to the free electrons in this phase would be more adequately defined in terms of an expression of the form $-(E^* + \frac{3}{2} kT_e)dN_e/dt$, where the term in the parentheses represents the average energy released to the free electrons for each electron recombination into the higher states. The value of E* would be approximately equal to the binding energy of the level immediately above the critical gap if de-excitation to lower levels by electron collision is negligible. It is interesting to solve for the electron temperature during this short period at the beginning of the relaxation process by simply equating this expression to the elastic en-

Table II.

1	time, t μsec	0	2	8	16	20	30	60	100	140
2	$-1.61\,(dN_{4P})/dt$ watts/cm^3	-	-	1.0	.38	.26	.11	.03	.02	.02
3	$-(V_o + \frac{3}{2}kT_e)\frac{dN_e}{dt}$ watts/cm^3	77.0	41.9	11.9	4.78	3.25	1.82	.58	.24	.14
4	$\dot{R}$ watts/cm^3	8.4	8.1	5.4	3.06	2.55	1.45	.64	.38	.32
5	σE^2 watts/cm^3	.63	.55	.39	.29	.27	.22	.14	.12	.10
6	$\dot{\Omega}$ watts/cm^3	69.2	34.4	7.9	2.39	1.43	.70	.11	0	-.02
7	$-(E^* + \frac{3}{2}kT_e)dN_e/dt$ watts/cm^3	21.4	11.7	3.3	1.33	.91	-	-	-	-
8	T_e^* °K	2850	2710	2480	2370	2330	-	-	-	-
9	N_e 10^{14}/cm^3	5.3	4.0	2.27	1.51	1.28	1.0	.59	.40	.30
10	T_e °K	>3500	>3500	≳3000	2580	2470	2370	2180	2100	2000
11	α 10^{-10} cm^3/sec	3.64	3.47	3.05	2.78	2.60	2.46	2.23	2.06	2.06

ergy loss term $\dot{\Omega}$.[21] If E* is chosen to be 0.94 ev, as calculated for the range of electron temperatures between 2000 and 3000° K, then calculated electron temperatures, T_e^*, fall somewhat lower than before, as shown in rows 8 and 10 in Table II. These temperatures look about right, since the steady state value before relaxation is about 3000 °K. This may be regarded as somewhat fortuitous, since it is not obvious that the remaining terms in the energy balance will exactly cancel under these conditions.

Finally, some speculative evidence that a quasi-steady state is not reached until about 20 microseconds is given by the light intensity data given in Figure 8. The 5P→4S transition decays quite rapidly at first, but reaches a low value by about 20 microseconds and decays slowly thereafter. The initial rapid decay would be expected in response to the initial drop in electron density, and the relatively slow variation indicates that after 20 microseconds the quasi-steady state may have been reached. The population of the 4P state does not begin to decay rapidly until after times ranging from 4 to 10 microseconds for various initial conditions. This indicates the relatively long times necessary for the 4P state to reach a quasi-steady state condition. This behavior is consistent with the fact that for these experiments, populations of the first excited state are not small compared to the density of free electrons, and that calculated de-excitation rates for these low-lying levels are not much faster than the rate of de-excitation across the critical gap.

Summary

As a result of the good agreement between experimentally determined values of electron temperature and electrical conductivity with values calculated from a two-temperature model, it can be concluded that the two-temperature model gives an accurate description of the plasma over a wide range of neutral gas temperatures, seed concentrations, and current density. At low current densities, the model is not adequate, and the primary sources of error appear to be the omission from the model of the atom-atom excitation process and the lack of thermal equilibrium between the electron gas and the populations of the excited states.

Measurements have been made of the electron-electron-ion recombination rate coefficients in potassium. The measured values have been compared with values calculated from a recombination rate theory employing the Gryzinski classical collision cross sections. The agreement between calculated and measured values is satisfactory.

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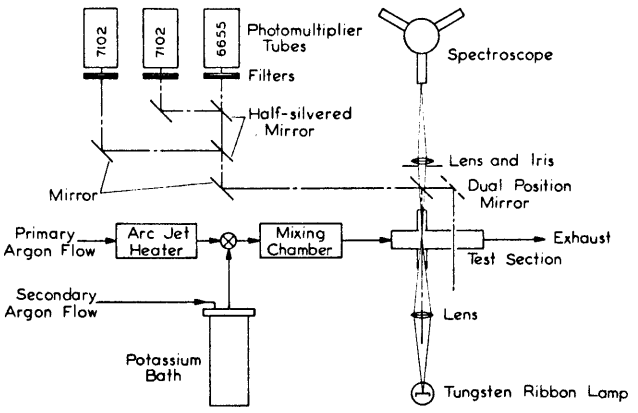

FIG. 1 SCHEMATIC DIAGRAM OF APPARATUS

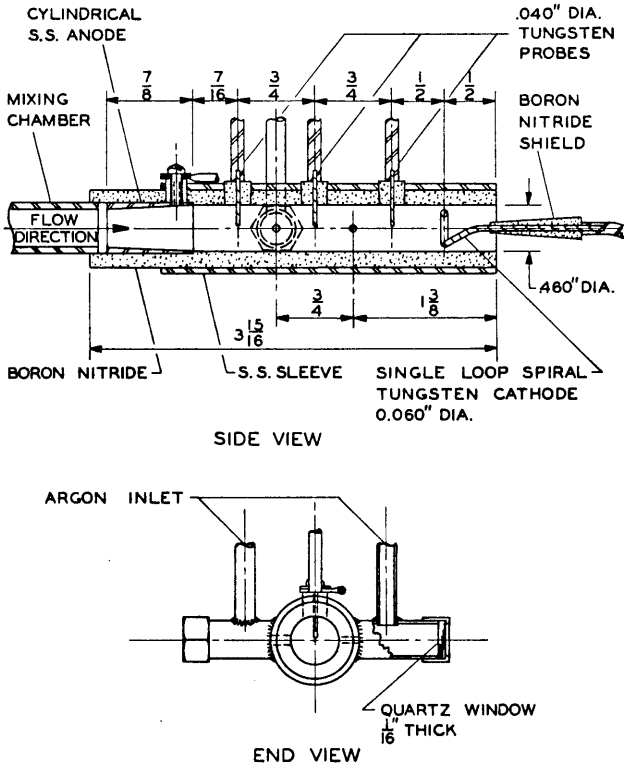

FIG. 2 SCHEMATIC DIAGRAM OF TEST SECTION

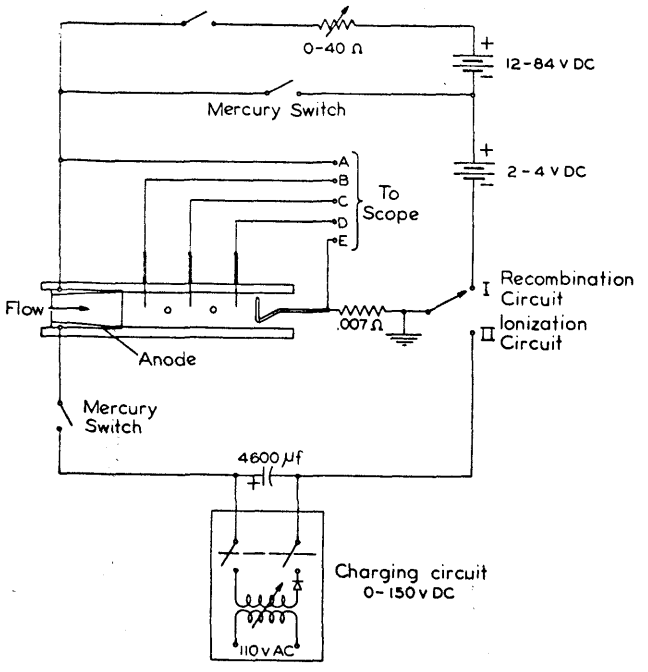

FIG.3 SCHEMATIC DIAGRAM OF ELECTRIC CIRCUIT

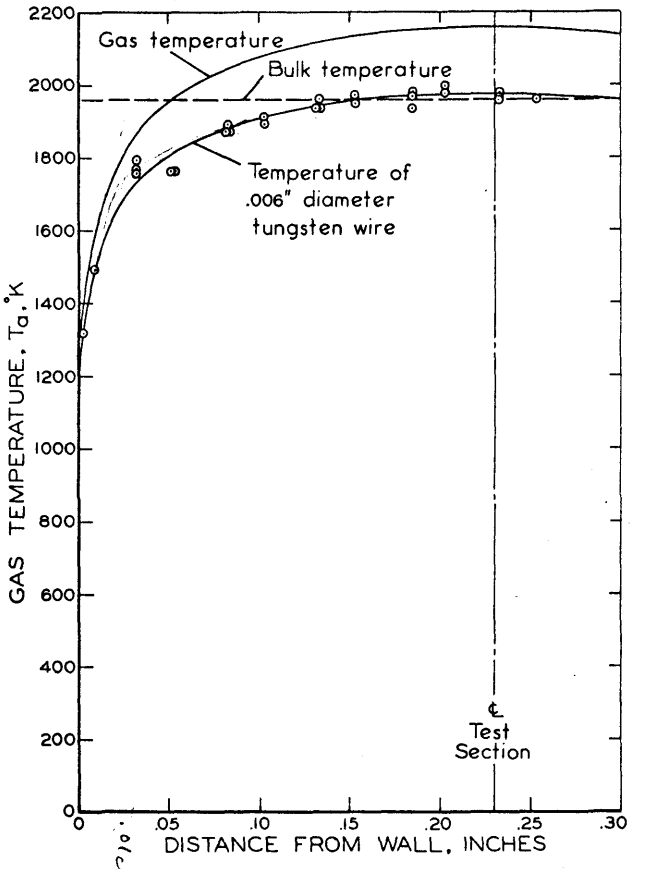

FIG. 4 TYPICAL TEMPERATURE DISTRIBUTION ACROSS TEST SECTION

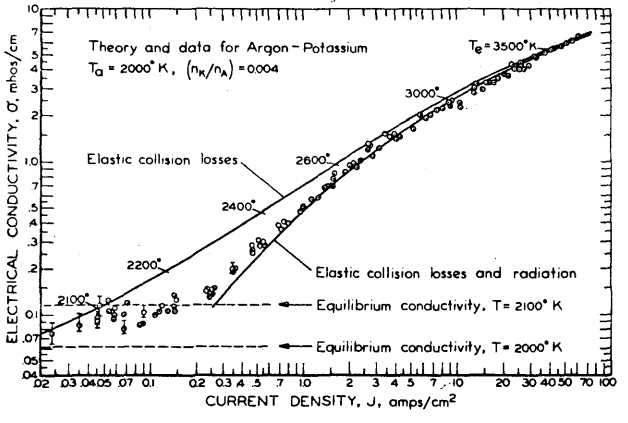

FIG.5 DEPENDENCE OF STEADY STATE CONDUCTIVITY ON CURRENT DENSITY

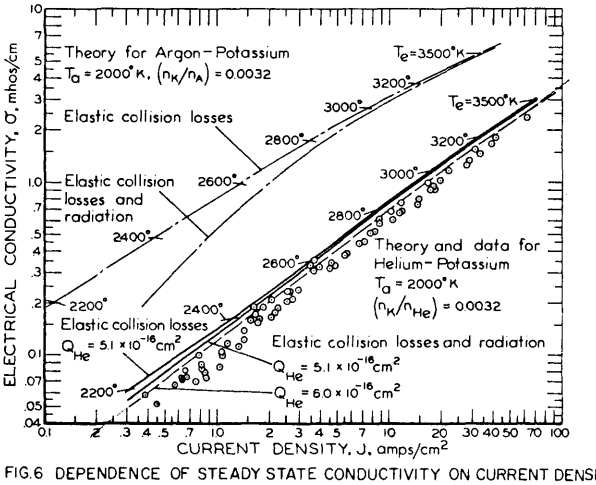

FIG.6 DEPENDENCE OF STEADY STATE CONDUCTIVITY ON CURRENT DENSITY

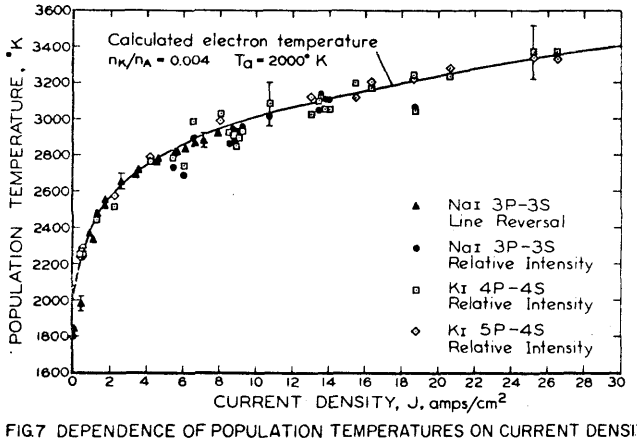

FIG.7 DEPENDENCE OF POPULATION TEMPERATURES ON CURRENT DENSITY

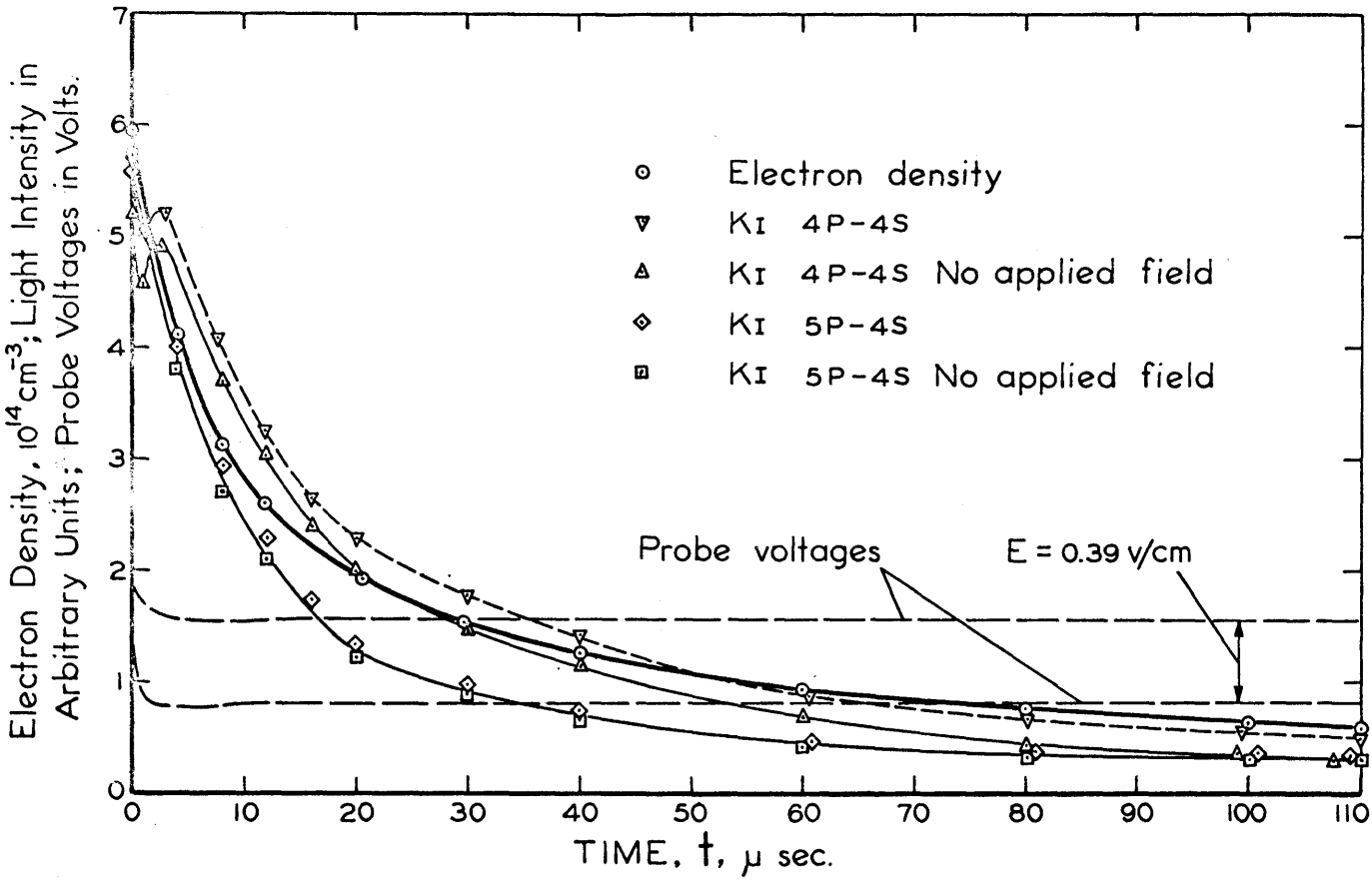

FIG. 8 TYPICAL RELAXATION DATA

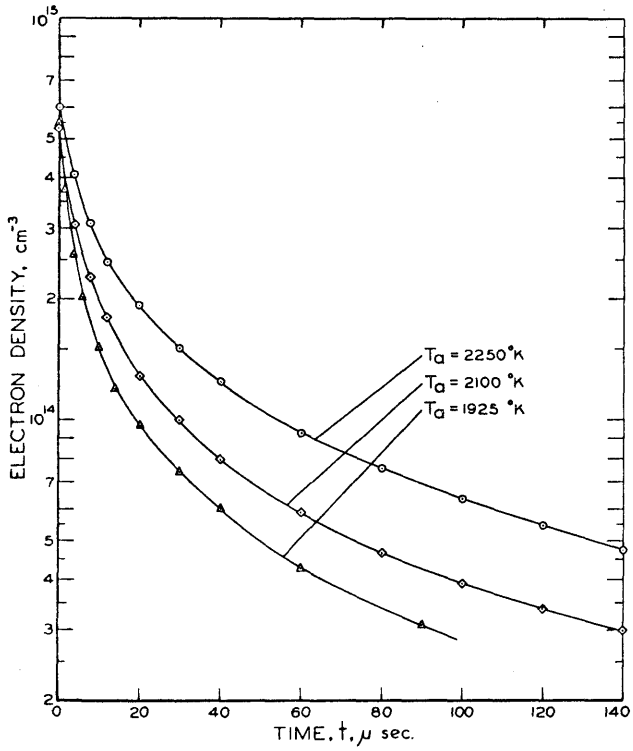

FIG. 9 TYPICAL VARIATION OF ELECTRON DENSITY DURING RELAXATION

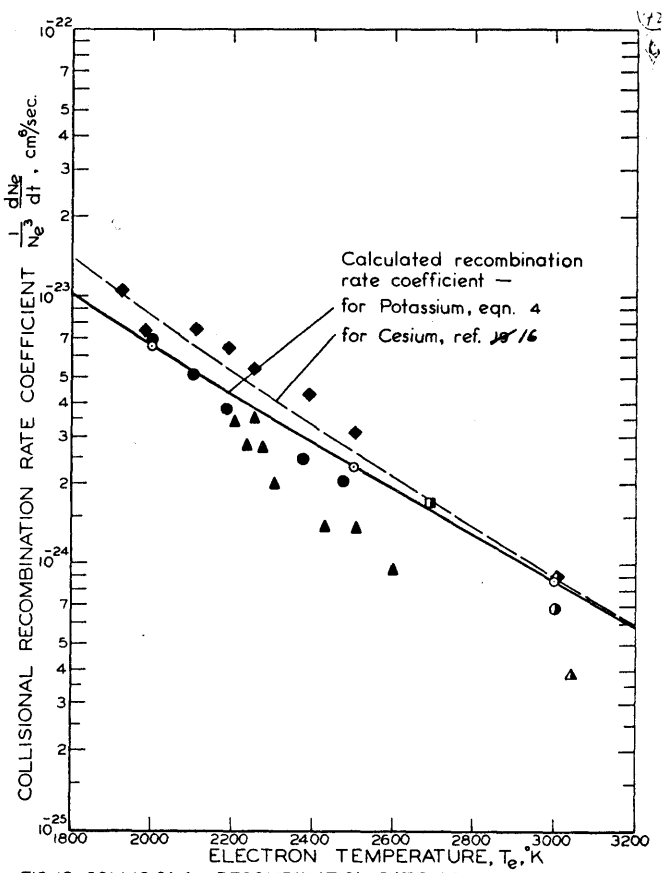

FIG. 10 COLLISIONAL RECOMBINATION RATE COEFFICIENT